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ABSTRACT OF THE DISCLOSURE

An assembly comprising a substrate designed for use as base stock for labels, tapes, bumper stickers, posters, etc., the substrate being coated with a water soluble, pressure sensitive adhesive composition, and a gas releasing reactant incorporated in the assembly so that upon activation thereof, the assembly self removes from the object to which it is adhered.

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As is known in the art, pressure sensitive adhesives comprise a class of adhesive compositions which share the common characteristic of being able to adhere to a surface by the application of nothing more than manual pressure. Adhesives of this type may be applied to various substrates such as paper, cloth and plastic films and the resulting coated substrates may then be converted to tapes, labels, posters, etc. which are useful for any number of applications.

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It is desirable that the adhesive composition be water soluble thereby offering the advantage of being removable from a substrate merely by washing with water. These adhesives avoid the use of organic solvents which, in addition to being costly, are often injurious to the user as well as to the substrates to which they are applied.

In washing used test tubes, soda bottles and the like, it is highly desirable that labels used thereon be capable of quick self removal so as to float off the object to which they are adhered within a brief period of time and without mechanical assistance. While water soluble, pressure sensitive adhesive labels self remove, manual assistance is normally required as

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wash solution contact alone is too time consuming.

BRIEF SUMMARY OF THE INVENTION

5 The present invention comprises a self removable, water soluble pressure sensitive adhesive coated sign or label stock assembly including a gas producing reactant. The gas producing reactant may be activated by immersion in water so as to self remove the sign or label from the object to which it is adhered by the driving force of gas bubbles formed between the adhesive and object.

10 The gas producing reactant may comprise a component or components which react on contact with water. In order to provide stability under highly humid conditions, it is preferred to use a two component system with one of the components in the adhesive and the other component in the wash solution or in a barrier layer between the adhesive and the sign or label stock. In 15 instances where the sign or label stock is formed of paper, one of the active ingredients may be incorporated in the paper or size coating during paper manufacture and the other component in the barrier or adhesive layer.

20 In order to entirely avoid the problem of providing a special wash solution and/or of high humidity inadvertently activating the gas releasing agent, a gas producing reactant which releases gas bubbles upon application of heat may be used. In this embodiment of the invention, self removal may be accom- 25 plished, for example, merely by raising the wash solution to the activating temperature.

30 In a still further embodiment of the invention, a water soluble barrier layer is used to separate the two components of the gas releasing systems and thus prevent inadvertent reaction therebetween. Upon immersion in water, the barrier layer dissolves and the two components react.

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DRAWINGS

FIGURE 1 is a cross section view of the present invention.

FIGURE 2 is a cross section of a modified form of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

Referring to FIGURE 1, a pressure sensitive adhesive label assembly 1 is shown, which is adapted to be printed and formed into labels. Assembly 1 includes base stock 3, barrier layer or size coating 5 and water soluble pressure sensitive adhesive coating 7. It is to be understood that while assembly 1 is referred to as a label, it is meant to represent any substrate to which pressure sensitive adhesives are conventionally applied, such as, tapes, posters, bumper stickers, etc.

The exposed surface of the water soluble pressure sensitive adhesive 7 is protected by a release sheet 9 which comprises a substrate 11 having a release coat 13 coated on the surface thereof adjacent the adhesive 7.

Base stock 3 may comprise cloth, plastic or any grade of paper, and generally, it is a highly finished book paper. Layer 5 may comprise a conventional size coating so as to provide hold out. In the alternative, layer 5 may comprise a barrier coating 5 adapted to prevent adhesive migration into the base stock.

The release sheet 9 is comprised of a paper substrate 11, preferably of the kraft type. Release coating 13, which is in contact with the pressure sensitive adhesive, may comprise any conventional material such as nitrocellulose, cellulose

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triacetate, silicone and any other conventional release material for pressure sensitive adhesives.

5 The water soluble pressure sensitive adhesive 7, may comprise any of those well known in the art, such as those described for example, in U. S. Patent 3,249,572 and U. S. Patent 2,985,609.

10 The above coatings are applied by any one of the well known methods such as roll coating, doctor blade coating, knife coating, etc., and dried in an oven at the proper temperature and lineal speed in accordance with practice well known in the art.

WATER ACTIVATED GAS RELEASING AGENT

15 In accordance with one embodiment of the present invention, a gas producing reactant is used which is adapted to be activated when immersed in water. The gas producing reactant may be included in adhesive layer 7, or, the reactant may, in whole or in part, be incorporated into other portions of the assembly, as hereinafter more particularly described.

20 The gas producing reactant may comprise a component or components which on contact with water, react to produce gas bubbles thereby driving the adhesive from the surface to which it is adhered. Examples of such materials are: saponin, calcium oxide, cesium carbonate, cesium tetroxide. Typical of  
25 a two component gas producing systems are the reactions of a mild acid such as formic, tartaric, citric, acetic, phosphoric, oxalic, titanilic or sulfamic with a metal salt such as sodium bicarbonate, sodium borate, sodium perborate, sodium acid pyrophosphate, sodium pyrophosphate peroxide, sodium citrate, sodium xylene sulfonate, sodium toluene sulfonate, lead carbonate,  
30 sodium aluminum phosphate, or sodium lauryl sulphate. With regard to the two component gas releasing systems, reference in

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the appended claims to a gas producing reactant, is meant to define one of the two components, and does not necessarily require the presence of all of the components of a plural component gas producing system.

5           While the above are given as illustrative of suitable gas releasing agents, other agents may be used, the prerequisite being that they be compatible with the material with which they are mixed, that they react within a short time period to meet the requirements of a particular application on contact with  
10           water, that they produce sufficient amountsof gas to accomplish the desired self removing function, and that the gas producing agents and their reaction products are non-toxic and non-hazardous to the degree required under particular condition of use. The acid reactant should not be added to an alkaline-medium while  
15           the salt should not be added to an acid medium.

#### LOCATION OF GAS PRODUCING REACTANT

          The gas releasing substance may be incorporated, in whole or part, into portions of assembly 1 other than the adhesive 7, as heretofore described. As an example, a two component  
20           gas releasing system may be used with one component located in base stock 3 or in barrier or sizing layer 5 and the other component in adhesive layer 7 so as to somewhat isolate the components to prevent inadvertent reaction under highly humid conditions. Still further one component may be located in barrier  
25           layer or sizing layer 5 and one component located in base stock 3.

          Referring to FIGURE 2, a modified form of the present invention is shown illustrating a further alternative, completely isolating the gas releasing components. In this embodiment, the  
30           two components of the gas releasing substance are separated and isolated by a barrier layer 21 with one component in base stock 19 or in sizing 23 and one component in pressure sensitive

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adhesive 25. Barrier layer 21 may be made water soluble so that upon immersion in water, the barrier 21 dissolves thereby allowing the gas producing reactants to rapidly react.

5       Such a barrier may be formed from materials that are water soluble film formers including substances from the following group and mixtures thereof: animal glues such as bone or hide glue, fish glue, casein, vegetable glues including natural gums such as gum tragacanth, gum dammer, gum karaza, agar agar, gum arabin; manufactured vegetable glues such as  
10       starch and starch derivatives including dextrin, British gum, potato dextrin, chlorinated starch; mineral glues including silicate of soda; and synthetic glues including polyvinyl alcohol; dextrin, carboxy, methyl cellulose and methyl and  
hydroxyethyl cellulose.

15       The gas producing reactant may be incorporated in a coating batch by mixing for a sufficient length of time to provide a well dispersed condition. In case of addition to the label stock, the reactant may be added to the pulp prior to formation on the wire.

20       The amount of gas producing reactants to be incorporated in the assembly will depend upon various factors, including the particular reactants used, and the speed desired for self removal. Speaking generally, the quantity of gas producing reactants will rarely, if ever, exceed 80% of the dry weight of the label and  
25       in most cases it will be substantially less than that amount.

      The following examples are typical of the invention incorporating a gas producing reactant. All references to parts are to parts by weight.



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EXAMPLE 1

5 6 parts of sodium bicarbonate were added slowly with mixing to 100 parts of a water soluble pressure sensitive adhesive. 14 parts of citric acid was then added slowly with continued mixing. 2 parts of a suspension agent, aluminum silicate, was then added and mixing was continued for 30 minutes until the components were thoroughly dispersed.

10 The batch was then coated on a one foot square release sheet of the type shown in FIGURE 1 and then dried in an oven for two minutes at 150 °F providing a 1 mil thick adhesive coating.

The coated release sheet was then cold laminated to label stock comprising a litho grade paper by passing the two assembled sheets through a two roll laminator.

15 The release sheet was then peeled off with the adhesive remaining on the label stock and one inch squares of the adhesive coated label stock were applied to a one and one-half (1½) inch square aluminum plates. Control samples having the same 1 mil thick water soluble pressure sensitive adhesive, but without the gas producing reactant, were applied to a second aluminum plate. The following illustrates the difference in self removal time when the above prepared plates were immersed in water at various temperatures.

	<u>Degree C</u> <u>Temperature</u>	<u>Self Removal Time</u> <u>For Adhesive Without</u> <u>Gas Releasing Agent</u>	<u>Self Removal Time</u> <u>For Adhesive With</u> <u>Gas Producing Reactant</u>
25	40 °	1810 seconds	100 seconds
	45 °	1650 "	90 "
	50 °	1505 "	75 "
	55 °	1415 "	65 "
	60 °	1210 "	55 "
30	65 °	1085 "	47 "
	70 °	935 "	38 "
	75 °	802 "	30 "
	80 °	650 "	23 "
	85 °	540 "	17 "

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From the above, it will be noted that self removal is increased by a factor about 20 at various temperatures by the addition of gas producing reactants.

EXAMPLE II

Label stock was prepared as in Example I in an identical manner and with identical formulation except that half the amounts of gas reactant were used in that 3 parts of sodium carbonate and 7 parts of citric acid were mixed into the adhesive. The stock was adhered to a plate as in Example I and immersed in water.

<u>Degree C</u> <u>Temperature</u>	<u>Self Removal Time</u> <u>For Adhesive With One</u> <u>Half Gas Producing</u> <u>Reactant of Example I</u>
40 °	145 seconds
45 °	125 "
50 °	84 "
60 °	83 "
65 °	73 "
70 °	52 "
75 °	69 "
80 °	32 "
85 °	26 "

EXAMPLE III

10 parts of sodium citrate were added slowly with mixing to 100 parts of a water soluble pressure sensitive adhesive. 3 parts of acetic acid was then added slowly with continued mixing for 30 minutes until the components were thoroughly dispersed.

The batch was then coated on a one foot square release sheet of the type shown in FIGURE 1 and then dried in an oven for two minutes at 150 °F. providing a 1 mil thick adhesive coating. The coated release sheet was then cold laminated to

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to label stock comprising a litho grade paper by passing the two assembled sheets through a two roll laminator.

The release sheet was then peeled off with the adhesive remaining on the label stock, and one inch squares of the adhesive coated label stock were applied to one and one-half (1½) inch square aluminum plates. The following gives the self removal time when the above prepared plates were immersed in water at various temperatures.

	<u>Degree C</u> <u>Temperature</u>	<u>Self Removal Time</u>
10	40 °	226 seconds
	45 °	202 "
	50 °	178 "
	55 °	164 "
15	60 °	150 "
	65 °	140 "
	70 °	125 "
	75 °	110 "
	80 °	91 "
20	85 °	77 "

EXAMPLE IV

12 parts of lead carbonate were added slowly with mixing to 100 parts of a water soluble pressure sensitive adhesive. 5 parts of titanitic acid was then added slowly with continued mixing for 30 minutes until the components were thoroughly dispersed.

The batch was then coated on a one foot square release sheet of the type shown in FIGURE 1 and then dried in an oven for two minutes at 150 °F providing a 1 mil thick adhesive coating. The coated release sheet was then cold laminated to label stock comprising a litho grade paper by passing the two assembled

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sheets through a two roll laminator.

The release sheet was then peeled off with the adhesive remaining on the label stock and one inch squares of the adhesive coated label stock were applied to one and one-half (1½) inch square aluminum plates. The following illustrates the difference in self removal time when the above prepared plates were immersed in water at various temperatures.

	<u>Degree C</u> <u>Temperature</u>	<u>Self Removal Time</u>
10	40 °	183 seconds
	45 °	167 "
	50 °	147 "
	55 °	138 "
	60 °	123 "
15	65 °	108 "
	70 °	92 "
	75 °	77 "
	80 °	60 "
	85 °	34 "

20

#### EXAMPLE V

6 parts of sodium bicarbonate were added slowly with mixing to 100 parts of a water soluble pressure sensitive adhesive and mixing was continued for 30 minutes until the components were thoroughly dispersed.

25

The batch was then coated on a one foot square release sheet of the type shown in FIGURE 1 and then dried in an oven for two minutes at 150 °F providing a 1 mil thick adhesive coating.

30

A barrier coating batch was then prepared by mixing 10 parts of polyvinyl alcohol with 90 parts of water. 56 parts of citric acid was then added with continued mixing until completely dispersed.

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The batch was then coated on a one foot square of label stock and dried providing a 1 mil thick barrier coating.

5 The coated release sheet was then cold laminated to the barrier coated label stock by passing the two assembled sheets through a two roll laminator.

10 The release sheet was then peeled off with the adhesive remaining on the label stock and one inch squares of the adhesive coated label stock were applied to one and one-half (1½) inch square aluminum plates. The following gives self removal time when the above prepared plates were immersed in water at various temperatures.

	<u>Degree C</u> <u>Temperature</u>	<u>Self Removal Time</u>
	40 °	136 seconds
15	45 °	120 "
	50 °	102 "
	55 °	75 "
	60 °	74 "
	65 °	48 "
20	70 °	39 "
	75 °	38 "
	80 °	34 "
	85 °	25 "

25 The above illustrates label stock having one gas producing reactant in the adhesive and the other gas producing reactant in the barrier coat. Due to isolation of the gas producing reactants, inadvertent reaction between the components tends to be minimized.

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EXAMPLE VI

10 parts of sodium carbonate were added slowly with mixing to 100 parts of a water soluble pressure sensitive adhesive. 14 parts of citric acid was then added slowly with continued mixing. 2 parts of a suspension agent, aluminum silicate, was then added and mixing was continued for 30 minutes until the components were thoroughly dispersed.

The batch was then coated on a one foot square release sheet of the type shown in FIGURE 1 and then dried in an oven for two minutes at 150 °F providing a 1 mil thick adhesive coating.

Label stock was prepared by adding 8 parts of  $\text{CaCO}_3$  to 100 parts of pulp comprising 50% hardwood kraft and 50 % softwood kraft. This slurry was then mixed for 15 minutes.

25 parts of a sizing agent consisting of an alkyline dimer together with .75 parts of a starch retention agent were then added to the slurry and mixed for 30 minutes. Label stock hand sheets were then formed by pouring the slurry on a 7 3/4 by 7 3/4 inch screen sheet mold. The sheet was then lifted off by use of a blotter sheet, put through a press and then placed on a steam drum to dry. The sheet so formed contained 5%  $\text{CaCO}_3$ .

A barrier coating batch was then prepared by mixing 90 parts of polyvinyl alcohol with 10 parts of water.

The barrier coating batch was then coated on the label stock and dried providing a 1 mil thick barrier coating.

The coated release sheet was then cold laminated to the barrier coated label stock by passing the two assembly coated sheets through a two roll laminator.

The release sheet was then peeled off with the adhesive remaining on the label stock and a one inch square of the adhesive coated label stock was applied to a one and one-half (1½)

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inch square aluminum plate. The following is self removal time when the above prepared plate was immersed in water at various temperatures.

5	<u>Temperature</u>	<u>Self Removal Time</u>
	40 °	159 seconds
	45 °	147 "
	50 °	116 "
	55 °	84 "
10	60 °	63 "
	65 °	53 "
	70 °	53 "
	75 °	43 "
	80 °	35 "
15	85 °	30 "

The above illustrates a two component gas releasing system with one component in the adhesive and one component in the label stock with a barrier sandwiched in between. The barrier separates the two components and prevents inadvertent reaction. This has advantage over the assembly in Example V as it provides complete isolation. In any coating operation containing dispersed particles, the particles during the drying of the coating will to some extent settle on the surface of the coating. Accordingly, the gas producing reactants in Example IV are only partially isolated. In this example, complete isolation is achieved.

#### CHEMICAL ACTIVATOR

In accordance with this embodiment of the present invention, components of the type selected under water activation, hereinbefore described, are employed except that one of the

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components is incorporated in the wash solution. This system is referred to as chemical activation as it is the introduction of the chemical component as well as water that initiates the reaction. Any of the two component gas releasers hereinbefore described may be used.

EXAMPLE VII

The following is typical of chemical activation in accordance with the present invention.

A water soluble pressure sensitive adhesive coated label specimen was prepared as in Example I except that the citric acid was omitted from the formulation.

A wash solution was then prepared by mixing 2 parts of acetic acid to 98 parts of water. Aluminum plates with the specimens thereon were immersed in a wash solution. The following gives the results.

<u>Temperature</u>	<u>Time for Self Removal</u>
40 °	130 seconds
45 °	124 "
50 °	98 "
55 °	80 "
60 °	74 "
65 °	65 "
70 °	45 "
75 °	37 "
80 °	29 "
85 °	26 "

This again illustrates a method for avoiding inadvertent reaction.



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HEAT ACTIVATION

In accordance with a further modification of the present invention, the gas producing reactant may be heat activated into a gas producing state. Such materials are conventionally referred to as blowing agents.

While this embodiment of the invention is not limited to any particular solid chemical gassing agent, the following have been found satisfactory for that purpose:

N, N' dimethyl N, N' dinitrosoterephthalamide; decomposed 95°C.

Aluminum Bromate; decomposes 212°.

Ammonium Bicarbonate; decomposes 140°F.

Ammonium Carbonate; decomposes 136.4°F.

Sodium Carbonate; decomposes 95°F.

Sodium Ammonium Phosphate; decomposes 174°F.

Cadium Phosphate; monobasic decomposes 212°F.

While the above are given as illustrative of suitable heat responsive gassing agents, other such agents may be used, the prerequisite being that they be solid at room temperature, decompose within the range of aqueous washing solution 90°F to 212°F. produce a sufficient amount of gas to drive off the stock from the adhered to surface and that both the gassing agent and decomposition products be non-toxic and non-hazardous to handle. The decomposition temperature range may be expanded to include other materials as under certain conditions normal use of the label or sign would be limited to a small range. For example, freezer labels are subject to a temperature range of -30° to 40°F. Accordingly, blowing agents which decompose above 40°F may be used.

The following is a typical example of a heat activated self removing adhesive.

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EXAMPLE VIII

5        **A** To 100 parts by weight of water soluble pressure sensitive adhesive is added <sup>10</sup>parts of N, N' dimethyl N, N' dinitrosoterephthalamide (30% white mineral oil). The batch was then applied on a one foot square highly finished litho paper and dried providing a 1 mil thick coating of adhesive. A one inch square of this sheet was applied to a one and one-half (1½) inch square aluminum plate. The square self removed in 120 seconds when immersed in water at 85°C.

10                The foregoing is considered illustrative only of the principles of the invention. Since numerous modications and changes will readily occur to those skilled in the art, it is not desired to limit the invention to the exact construction and operation shown and described, and accordingly all suitable  
15        modifications and equivalents may be resorted to as falls within the scope of the invention.

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THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. An assembly consisting essentially of at least two layers of material, one of said layers including a pressure sensitive adhesive for adhering said layers to a substrate, at least one of said layers including: a gas producing reactant capable of reacting so as to produce gas so that the assembly tends to be driven by the force of the released gas from a substrate, to which it is adhered.
2. An assembly as defined by claim 1, said pressure sensitive adhesive layer being coated on the other of said layers, said pressure sensitive adhesive including said gas producing reactant.
3. An assembly as defined by claim 1, said pressure sensitive adhesive layer being coated on the other of said layers, said other layer including said gas producing reactant.
4. An assembly as defined by claim 1, at least one material sandwiched between said pressure sensitive adhesive layer and the other of said layers, said other layer including said gas producing reactant.
5. An assembly as defined by claim 2, said gas producing reactant being stable under normal ambient conditions, said gas producing reactant being capable of reacting with water to produce said gas.
6. An assembly as defined by claim 3, said gas producing reactant being stable under normal ambient conditions, said gas producing reactant being capable of reacting with water to produce said gas.
7. An assembly as defined by claim 4, said gas producing reactant being stable under normal ambient conditions, said gas producing reactant being capable of reacting with water to produce said gas.

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(8) An assembly as defined by claim 2, said gas producing reactant being capable of reacting with a second component in the presence of water to produce said gas.

(9) An assembly as defined by claim 3, said gas producing reactant being capable of reacting with a second component in the presence of water to produce said gas.

(10) An assembly as defined by claim 4, said gas producing reactant being capable of reacting with a second component in the presence of water to produce said gas.

(11) An assembly as defined by claim 2, said gas producing reactant comprises a blowing agent which when heated to a critical temperature decomposes with an evolution of gas.

(12) An assembly as defined by claim 3, said gas producing reactant comprises a blowing agent which when heated to a critical temperature decomposes with an evolution of gas.

(13) An assembly as defined by claim 4, said gas producing reactant comprises a blowing agent which when heated to a critical temperature decomposes with an evolution of gas.

(14) An assembly as defined by claim 4, said material being water soluble.



FIG. 1

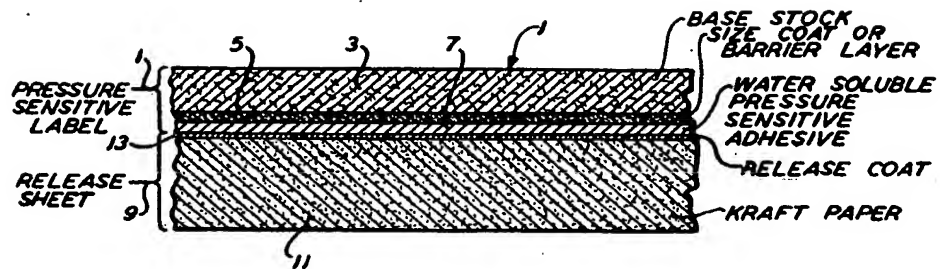
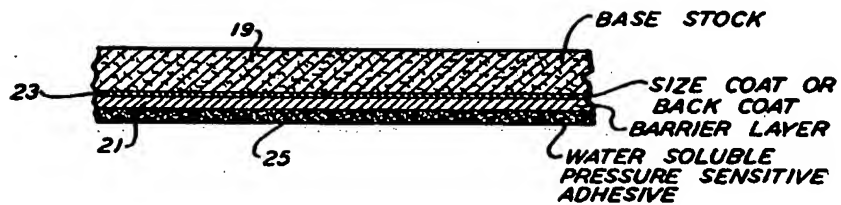


FIG. 2



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PATENT AGENT

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